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PROBLEMS WITH THE USE OF MICOTINE AS A PREDICTIVE ENVIRONMENTAL TOBACCO SMOKE MARKER

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A series of experiments was performed to evaluate the utility of nicotine as an environmental tobacco smoke (ETS) marker. Two University of Kentucky reference eigarettes (1R4F) were smoked in an 18-m<sup>2</sup> environmental chamber. Air exchange rates within the chamber were varied from 0-4 air changes per hour, and the concentrations of numerous ETS components were monitored for up to six hours after smoking. Under most ventilation conditions, nicotine initially decayed more rapidly than other ETS constituents; however, as sampling time was extended, nicotine decayed more slowly. The change in nicotine decay rate can lead to overestimation of ETS exposure when nicotine is used as the sole ETS marker. Confirmatory results obtained from other field and chamber studies are also presented.

### INTRODUCTION

The utility of nicotine as an environmental tobacco smoke (ETS) marker has been questioned by a number of investigators (1-3). Some have simply stated that it is a poor marker (1), while others have suggested that nicotine may underestimate ETS exposure (2,3) by as much as an order of magnitude.

The National Research Council (NRC) recommended that any chosen ETS marker should be present in a consistent ratio to ETS components of interest (4). To attain this criterion, the marker should possess the same decay characteristics as the component of interest.

If an ETS component is not generated or eliminated by chemical reaction, or does not interact with environmental surfaces, then it should demonstrate a first order decay with a rate constant proportional to the air exchange rate in a microenvironment. Previous studies have shown that nicotine does not undergo first order decay in microenvironments (2,3).

This present work was performed to systematically evaluate the effect of meetine's unique decay on its ratio to other ETS components. Due to nicotine's different decay rate, both ventilation rate and sampling time would be expected to exert an effect on the ratio of nicotine to other ETS constituents. Therefore, ratios were determined at air exchange rates of 0, 0.5, 1, 2 and 4 air changes per hour (ACH), and time-weighted-average concentrations of selected analytes were determined for 0.30, 0.60, 0.120, 0.240, and 0.360 minutes following the smoking of 2 University of Kentucky reference eigarettes (1R4F). The results obtained in the chamber were then related to results obtained previously in field and chamber studies.

### EXPERIMENTAL

# Chamber Studies

All ETS decay experiments were performed in an 18-m<sup>3</sup> environmental chamber described elsewhere (5). Three to five replicate experiments were performed at 0, 0.5, 1, 2, and 4 ACH. Real-time nicotine concentrations were monitored with a SCIEX TAGA 6000 tandem mass spectrometer (6). Real-time concentrations of carbon monoxide, nitrogen oxides, volatile organic compounds (estimated by FID response), and particle mass concentration were obtained with commercial analyzers described elsewhere (7). Vapor phase alcotine and 3-ethenylpyridine were collected using XAD-4 sorbent tubes and analyzed by gas chromatography with nitrogen phosphorus detection (8). Solanesol, gravimetric respirable suspended particles (RSP), ultraviolet particulate matter (UVPM), and fluorescent particulate matter (FPM) were collected on Fluoropore filters and analyzed as described elsewhere (9,10). Duplicate nicotine and particulate samples were collected over the periods 0-30, 30-60, 60-120, 120-240, and 240-360 minutes, and integrated average concentrations were determined.

Each run during the decay sate studies lasted a total of 384 minutes. The final twelve minutes of the run were used to measure background concentrations of ETS constituents. A smoker then entered the chamber and smoked two University of Kentucky 1R4F cigarettes in 10½ minutes. The two cigarettes were lit at 30-second intervals, and the smoker took one puff on alternating cigarettes at one-minute intervals. Each cigarette lasted for an average of ten puffs. At 24 minutes, the smoker exited the chamber which was subsequently resealed for the final 360 minutes of the experiment.

The effect of residual nicotine on a smokens' clothing was determined from the average of 15 runs performed at various times over a three-month period in the environmental chamber which was operated at 0 ACH. After a twelve-minute background measurement, the smoker entered the chamber and stayed for twelve minutes. The smoker then exited the chamber, and nicotine concentration was monitored for an additional 36 minutes.

### Field Studies

Nicotine samples were collected on XAD-4 sorbent tubes and analyzed by the method of Ogden et al. (8). Six-hour samples were collected in a smoker's van and the den of a non-smoker's house. Eight one-hour samples were collected overnight in a B767 aircraft which had completed a flight on which smoking was allowed.

Particulate samples were collected and analyzed for the aircraft study by the same methods used in the chamber study.

#### RESULTS AND DISCUSSION

## Nicotine Decay

At each of the air exchange rates studied, nicotine initially decayed more rapidly than the other ETS constituents measured. However, at longer times the decay of nicotine slowed, and typically achieved a near-steady state concentration which was higher than the initial background level. On the other hand, constituents such as CO, volatile organic compounds, nitrogen exides, and particle mass concentration decayed according to first order kinetics with decay rate constants which were proportional to the air exchange rate. The other constituents did decay to initial background levels in a time-scale consistent with normal first order decay.

A plot of nicotine concentration <u>vs.</u> time obtained from the average of five runs performed at 2 ACH is shown in Figure 1. The solid line in this figure is the average nicotine concentration measured in the chamber. The dashed line in the same figure is the nicotine concentration profile which would be predicted by first order decay.

The theoretical curve in Figure 1 is representative of the behavior of measured other ETS constituents. Time-weightedaverage (TWA) concentration ratios between nicotine and the other constituents are proportional to the ratio of the areas under the two curves. For a sampling period extending from 0-30 minutes, the ratio of nicotine to analyte will be lower than that predicted by first order kinetics. At about 30 minutes, nicotine decays less rapidly than the other constituents, and for sampling times greater than 60 minutes; the ratio of nicotine to

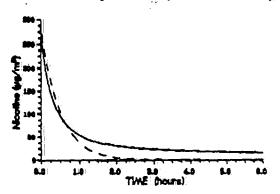


Figure 1. Average of five real-time nicotine concentrations (solid) measured in a controlled environment chamber operated at two ACH. The dashed line represents the concentration predicted by a first order decay mechanism.

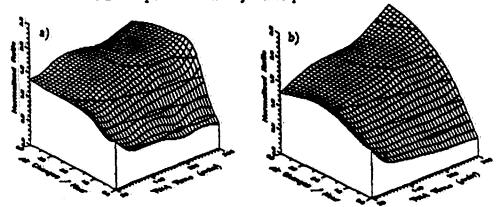
other analytes will become increasingly larger than those predicted by first order decay of constituents. The magnitude of overestimation becomes larger if measurements are started at times long after smoking has occurred. For the example illustrated in Figure 1, the other constituents would have decayed to background at about 180 minutes; but significant concentrations of nicotine are still present in the chamber. In this case, a person exposed to the atmosphere in the chamber would not be exposed to measurable ETS particulate, CO, or volatile organic compounds, but they would have measurable exposure to nicotine.

### Ratios Involving Nicotine

The actual variations of nicotine to RSP and nicotine to FID response ratios as a function of both sampling time and air exchange rate are shown in Figures 2a

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and 2b. Ratios presented in these figures have been normalized to the values obtained for the 0430 minute sample at 0 ACH. Sampling for increasing periods of time or sampling for a constant period of time at different air exchange rates has a dramatic influence on the observed ratio of nicotine to either vapor or particulate phase ETS components. Furthermore, these figures demonstrate that ratios of nicotine to other ETS components which are determined in chambers operated in static modes are not applicable to other environments because of the large overestimation of ETS exposure which they would predict.



Figures 2s & 2h. Normalized ratios between nicotine and gravimetric RSP (2a) and FID response (2b) as a function of both air exchange rate and sampling time. All values in figure 2a and 2b have been normalized to the ratio at 0 ACH and 30 min (0.108 in 2a & 74.1 in 2b). Lines of constant ratio are drawn at intervals of 0.333 beginning at a ratio of 0.667.

## Field Measurements

Results consistent with those obtained in the chamber also have been observed in the field. Background nicotine concentrations in the absence of smoking have been measured in homes, automobiles and aircraft. In addition, nicotine desorption from the clothes and person of a smoker has been observed in the environmental chamber. Results of these studies suggest that the initial rapid decay of nicotine is due to its adsorption on clothing and other surfaces. As atmospheric nicotine is depleted, due to the effects of adsorption and dilution by fresh air, adsorbed nicotine then desorbs, and leads to measurable nicotine concentrations in the absence of smoking.

Figure 3 shows that smokers can be a source of nicotine contributed to the environment even when they are not smoking. This figure shows the average increase in nicotine concentration measured during 15 replicate experiments performed over a three-month period in which a smoker entered the environmental chamber for twelve minutes. The increased nicotine concentration is presumably due to the evolution of motine from the smoker's clothes. When the smoker exited the chamber, the nicotine concentration ceased its increase.

A nicotine concentration of 0.09  $\mu g/m^3$  was measured in the den (=45 m<sup>3</sup>) of a non-smoker's house. The sample was obtained two days after smoking had occurred in the room. This background level corresponds to small amounts of residual nicotine desorbing from room furnishings over a long period of time.

Nicotine samples were collected in a van (=5 m<sup>3</sup>) in which smoking regularly

occurred. The sample was taken overnight and at least four hours after smoking had occurred in the vehicle. The 28W obtained sample dashboard level =40 cm above an open ashtray. A background concentration of 0.126 µg/m<sup>3</sup> was measured in the vehicle. Once again, this background is not due to the presence of ETS, but instead it comes from nicotine desorbing from the interior of the van and from cigarette butts present in the achtray.

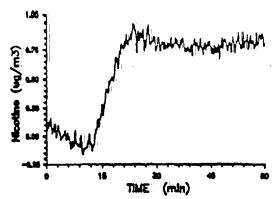


Figure 3. Average increase in nicotine concentration due to the nicotine desorption from a smoker's clothing in a controlled environment test chamber operated at 0 ACH. The smoker was in the chamber for the period 12-24 minutes.

Samples were also collected in a B767 aircraft in which smoking was permitted. The aircraft had returned from a regularly scheduled 4 hour 20 minute flight from Los Angeles, CA to Charlotte, NC. Samples were collected when the plane was on the ground and parked at the gate after all passengers and crew had left the plane. While the samples were collected, the aircraft was served by an auxiliary heating, ventilating and air conditioning system which provided fresh air at 13-26 ACH (11). Smoking was not permitted while the aircraft was at the gate, and none was observed. Samples were obtained in the coach smoking section and seats in the non-smoking border section. A total of 64 cigarette butts were counted in the ashtrays prior to sampling. The results of this investigation are presented in Table I. During time

period 4, the crew collected trash, emptied ashtrays during period 5, and vacuumed the cabin during period 7. UVPM measurements obtained from samples taken over the entire 8-hour period showed less than 1.5 μg/m<sup>3</sup> particulate matter was present which could possibly be attributed to ETS. Once again, the only source of nicotine in the cabin would appear to be nicotine desorbing from interior surfaces

Table I: Background nicotine concentrations (µg/m³) measured overnight on a B767 aircraft with audilary ventilation. Smoking samples (S.) were obtained in the coach smoking section. Non-smoking samples (N.S.) were obtained in coach smoking/nonsmoking border seats.

		Nicotine	
Sample	Time	S.	<u>N.S.</u>
1.	22:20-23:20	5.2	1.7
2	23:20-00:20	4.9	13
3	00:20-01:20	2.8	1.1
4	01:20-02:20	8.7	1.8
5	02:20-03:20	8.0	1.6
6:	03:20-04:20	4.7	1.3
7	04:20-05:20	5.2	2.7
8	05:20-06:20	8.2	2.5

and digarette butts. Measurable nicotine exposure could be expected in the non-smoking boundary section without concurrent exposure to ETS gas or particulate phase material.

## CONCLUSIONS

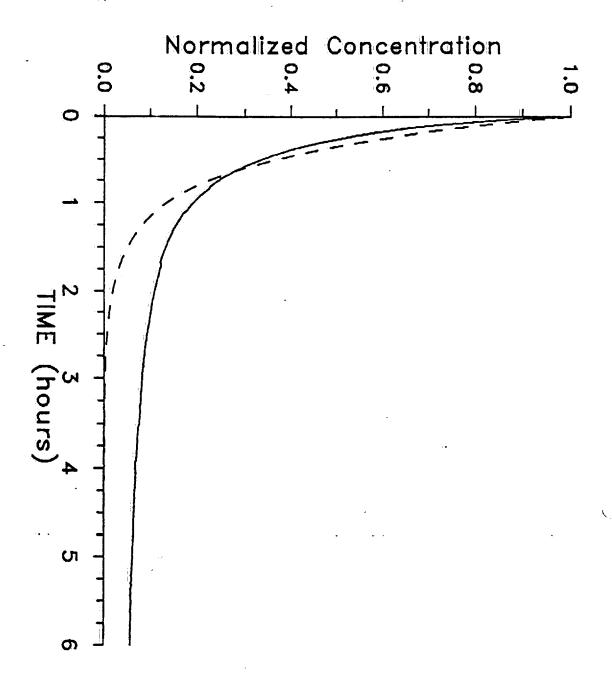
Nicotine does not fit the NRC criteria for an ETS marker. The ratio of nicotine to other ETS constituents such as RSP are highly variable and dependent on both the air exchange rate at the sampling site and sampling time. Desorption of nicotine from clothing, interior surfaces, and cigarette butts leads to measurable nicotine exposure in the absence of ETS. The findings reported here show that when nicotine is used as the sole marker, it may greatly overestimate ETS exposure:

#### ACKNOWLEDGEMENTS

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Nelson et al. Figure 1.